

Supporting Information

Directly Coating Multifunctional Interlayer on the Cathode via Electrospinning for Advanced Lithium Sulfur Batteries

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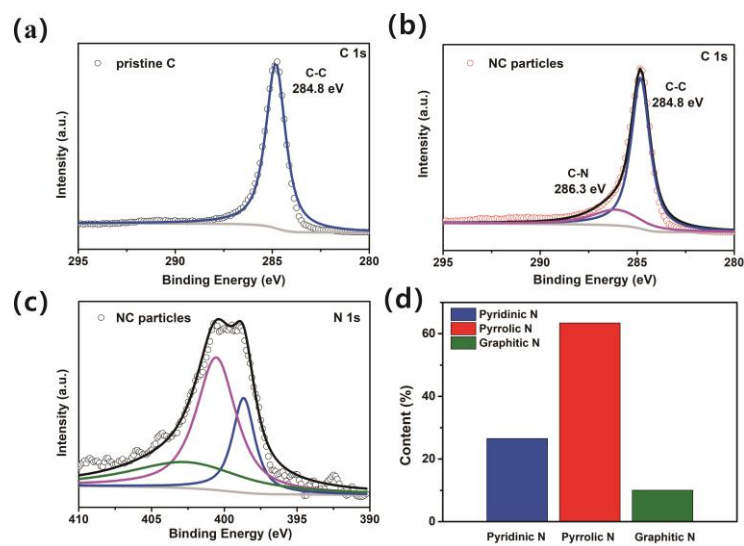


Figure S1. (a) C 1s spectra of pristine C, (b) C1s spectra of NC particles, (c) N 1s XPS spectrum of NC particles, (d) corresponding N content.

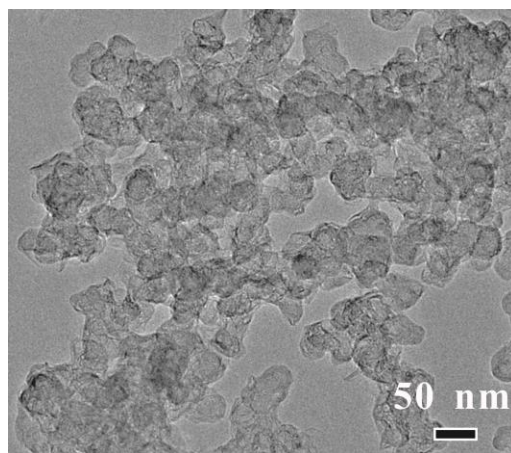


Figure S2. TEM image of NC particles.

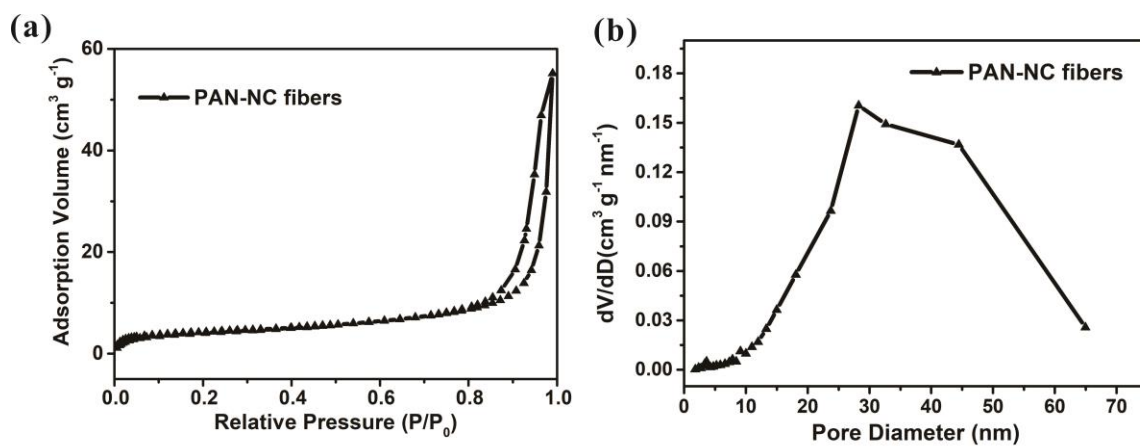


Figure S3. (a) nitrogen adsorption-desorption isotherm of PAN-NC fibers, (b) corresponding pore size distribution.

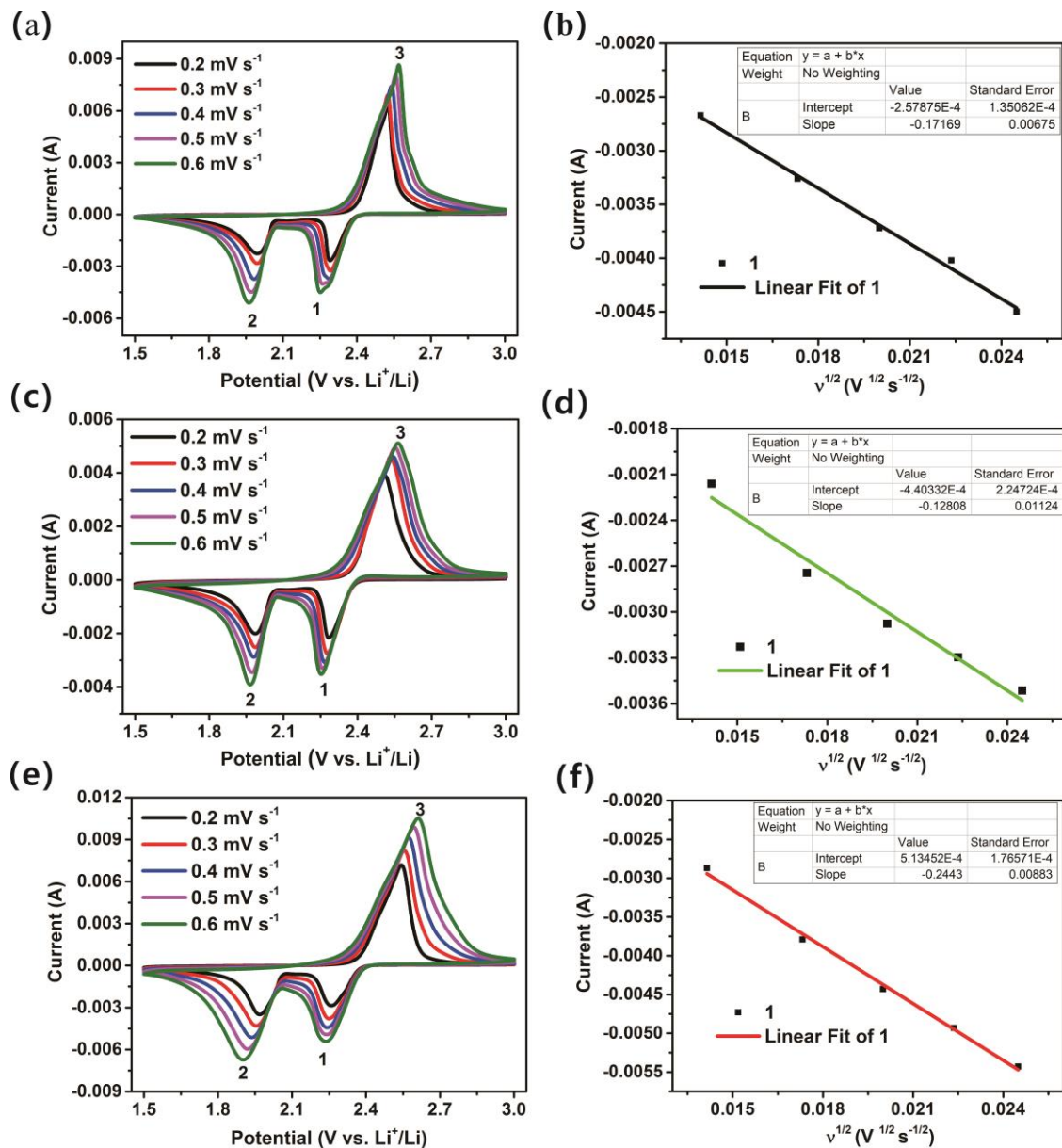


Figure S4. CV curves at different scanning rates and corresponding $i_{pa} - v^{1/2}$ scatters and linear fitting of (a, b) sulfur cathode, (c, d) PAN@cathode, (e, f) PAN-N@Cathode.

The diffusion coefficient of Li^+ (D_{Li^+}) can be calculated from the following Randles-Sevcik equation:¹⁻²

$$I_p = 0.4463nFAC\left(\frac{nFvD}{RT}\right)^{1/2} \quad (1)$$

Where I_p is the peak current (A), n is the number of electrons transferred, F is the Faraday's constant (96485 C mol^{-1}), A is electrode area (1.14 cm^2), C represents concentration of Li^+ (mol cm^{-3}), R is gas constant ($8.314 \text{ J K}^{-1} \text{ mol}^{-1}$), T is temperature (K), D is Li^+ diffusion coefficient ($\text{cm}^2 \text{ s}^{-1}$), v is scan rate (V s^{-1}). Based on the equation 1, the value of D can be obtained from the slope of $dI/dv^{1/2}$. Peak 1 in CV plots is chosen for calculation.

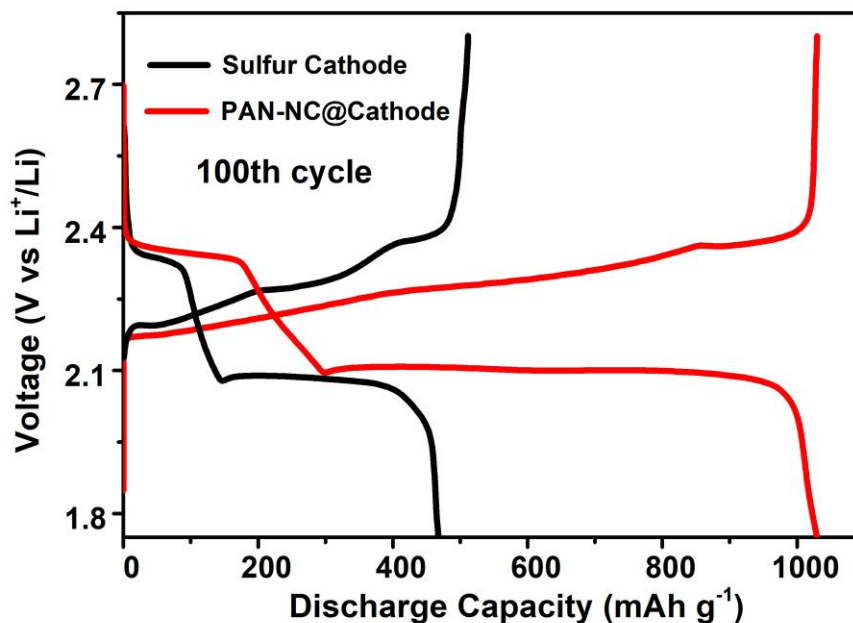


Figure S5. Discharge and charge plots of sulfur cathode and PAN-NC@Cathode at 200 mA g^{-1} after 100 cycles.

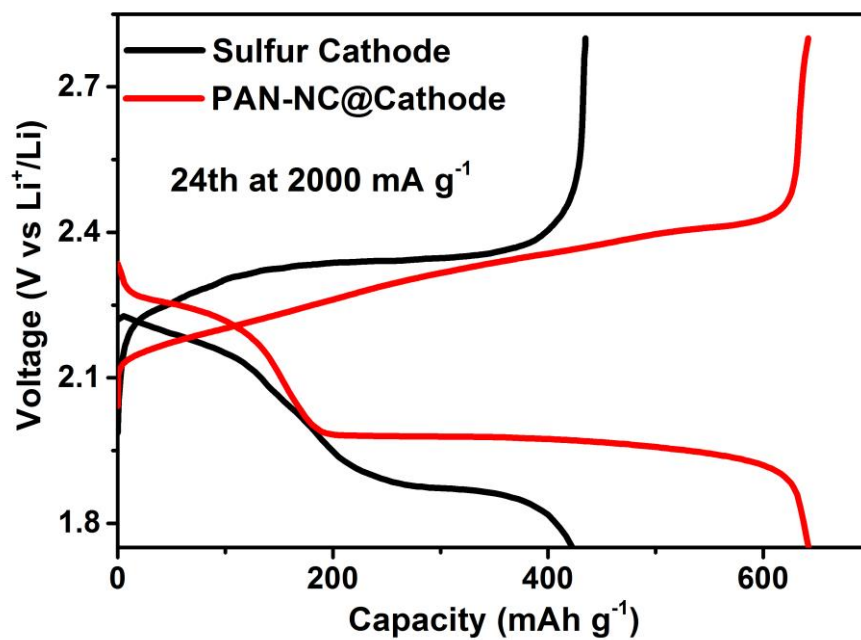


Figure S6. Discharge and charge plots of sulfur cathode and PAN-NC@Cathode at 2000 mA g⁻¹ at the 24 cycle.

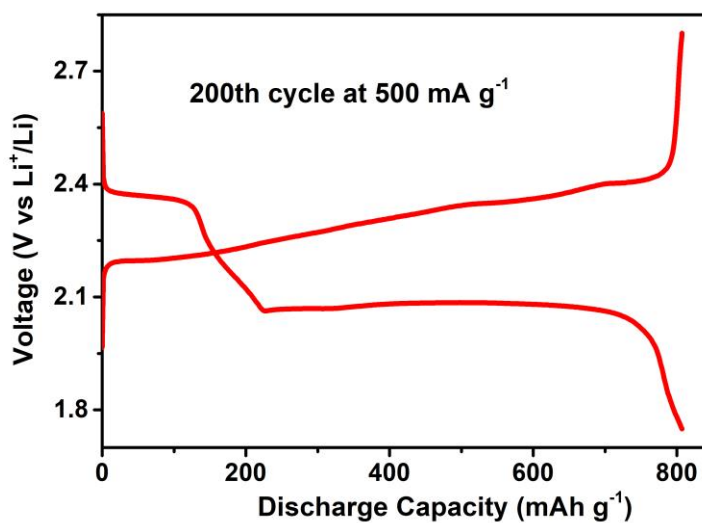


Figure S7. Discharge and charge plots of PAN-NC@Cathode at 500 mA g⁻¹ after 200 cycling.

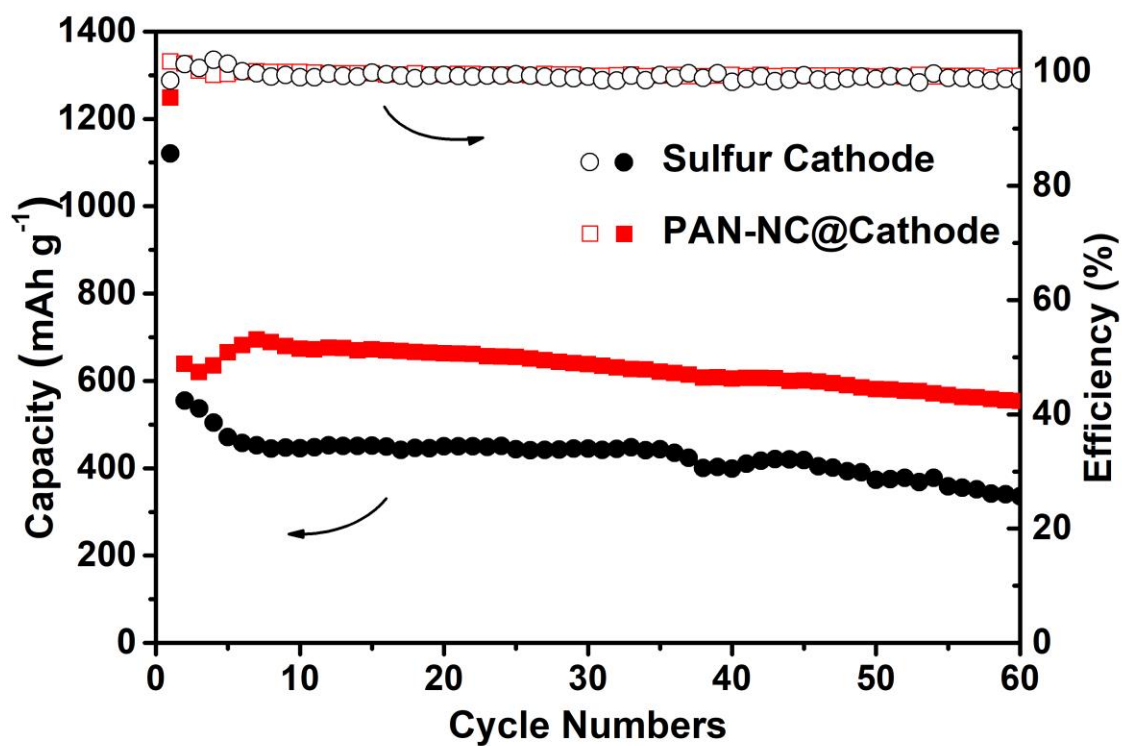


Figure S8. The cycling performances of sulfur cathode and PAN-NC@Cathode at 2000 mA g^{-1} after one cycle activation at 200 mA g^{-1} .

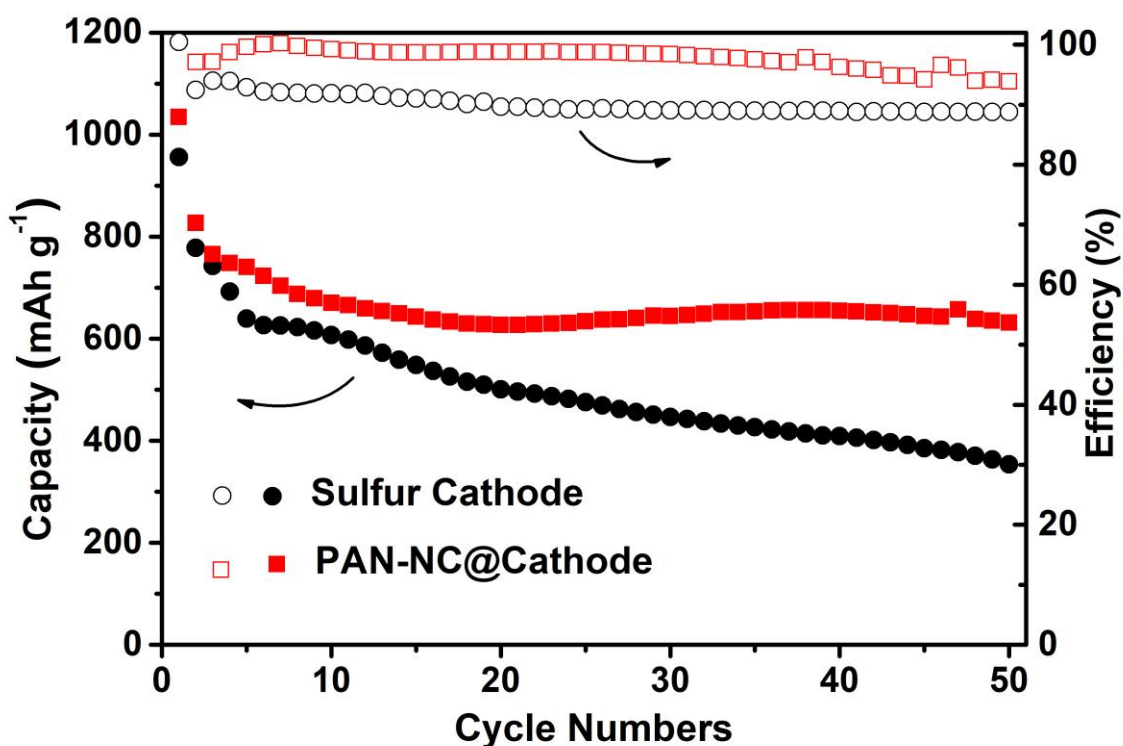


Figure S9. The cycling performances of sulfur cathode and PAN-NC@Cathode at 200 mA g^{-1} with higher sulfur loading of 4 mg cm^{-2} .

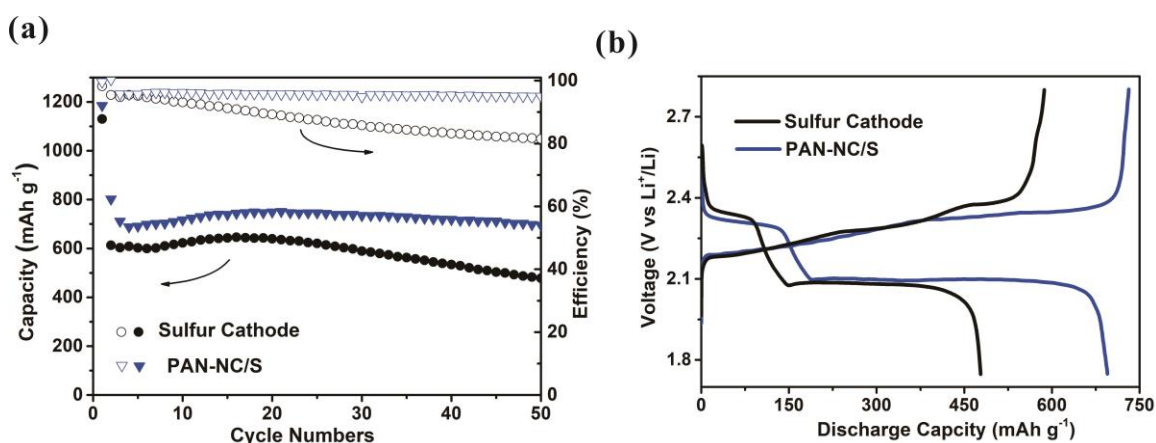


Figure S10. (a) the cycling performances of sulfur and PAN-NC/S cathodes at 500 mA g^{-1} after initial activation at 200 mA g^{-1} , (b) the discharge and charge curves of sulfur and PAN-NC/S cathodes at the 50th cycle.

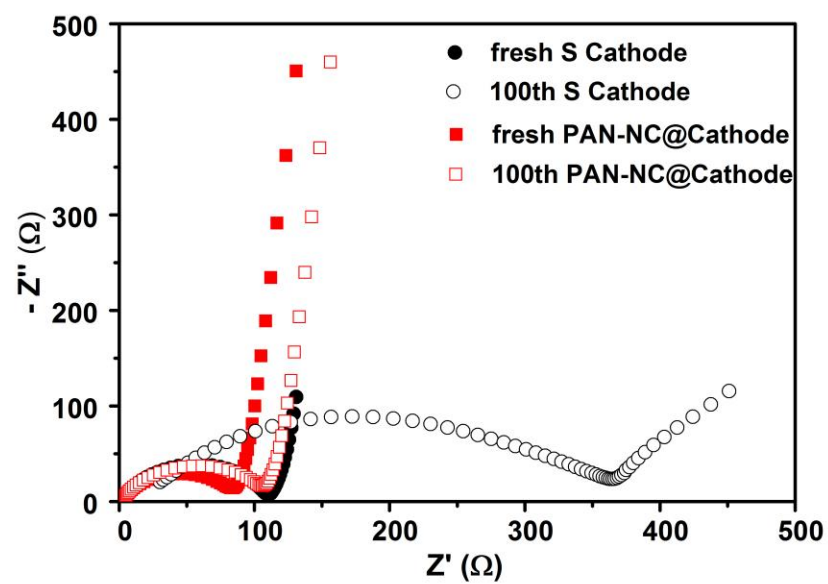


Figure S11. EIS spectra of cathodes before and after cycling

Table S1. The references³⁻¹⁰ about adding interlayer in Li-S batteries.

	interlayer	method	Weight (mg cm ⁻²)	cathode	Current density (mA g ⁻¹)	Capacity (mAh g ⁻¹) after 100	Capacity retention
Inserting	³ Microporous carbon paper (MCP)	1 mixing conductive carbon with binder at 3:2 mass ratio	-	S	1675	1000	85.0%
		2 roll-pressing					
	⁴ Reduced graphene oxide with carbon black (rGO-CB)	1 suction-filtering rGO-CB suspensions	-	S	200	894	70.9%
		2 heat treatment					
	⁵ tubular polypyrrole (T-PPy) film	1 self-degraded template method	1	S/C	335	~964	68.9%
		2 vacuum filtration and drying					
	⁶ Cyclized-Polyacrylonitrile Modified CNF (CP@CNF)	1 electrospun CNF from PAN	-	S	502	910	85.1%
		2 dip-coated PAN, thermal treatment					
	⁷ TiO ₂ decorated CNF (CNF-T)	1 electrospun CNF from PAN	0.5–0.6	S	335	~840	63.2%
		2 dip-coated TiO ₂ , thermal treatment					
coating	⁸ V ₂ O ₅ -Decorated Carbon Nanofiber	1 electrospun CNF from PAN	1	S	500	889	87.1% ^b
		2 solvothermal , thermal treatment					
	⁹ TiO ₂ /Graphene	coating with the doctor blade	0.1	PCNTs-S	837	~1040 after 300	99% ^a
This work	¹⁰ poly (3,4 ethylenedioxythiophene)-poly (styrene sulfonate)/Black Pearl (PS/BP)	electrospinning	0.09	NPCS-S	502	920	88.5%
	PAN-NC	electrospinning	0.14	S	200	1029	80.5%
					500	840	90.9% ^b

^a with a gradual increase for the first 20 cycles, ^b after one cycle activation, the capacity retention is calculated based on the second capacity.

Table S2. The results of resistivity measurements.

materials	ρ Ω cm	σ s m ⁻¹
PAN	14265	7.0×10^{-3}
Carbon black	0.1	1.0×10^3
PAN-NC	0.3	3.3×10^2

Electrical resistivity was measured with a four-contact method under the pressure of 4 MPa.

References

1. Li, H.; Jiang, J.; Wang, F.; Huang, J.; Wang, Y.; Zhang, Y.; Zhao, J., Facile Synthesis of Rod-like Cu_{2-x}Se and Insight into its Improved Lithium-Storage Property. *ChemSusChem* **2017**, 2235-2241.
2. You, Y.; Yao, H. R.; Xin, S.; Yin, Y. X.; Zuo, T. T.; Yang, C. P.; Guo, Y. G.; Cui, Y.; Wan, L. J.; Goodenough, J. B., Subzero-Temperature Cathode for a Sodium-Ion Battery. *Adv. Mater.* **2016**, 28, 7243-7248.
3. Su, Y. S.; Manthiram, A., Lithium-Sulphur Batteries with a Microporous Carbon Paper as a Bifunctional Interlayer. *Nat. Commun.* **2012**, 3, 1166-1172.
4. Wang, X.; Wang, Z.; Chen, L., Reduced Graphene Oxide Film as a Shuttle-Inhibiting Interlayer in a Lithium-Sulfur Battery. *J. Power Sources* **2013**, 242, 65-69.
5. Ma, G.; Wen, Z.; Wang, Q.; Shen, C.; Peng, P.; Jin, J.; Wu, X., Enhanced Performance of Lithium Sulfur Battery with Self-Assembly Polypyrrole Nanotube Film as the Functional Interlayer. *J. Power Sources* **2015**, 273, 511-516.
6. Li, Q.; Liu, M.; Qin, X.; Wu, J.; Han, W.; Liang, G.; Zhou, D.; He, Y.-B.; Li, B.; Kang, F., Cyclized-Polyacrylonitrile Modified Carbon Nanofiber Interlayers Enabling Strong Trapping of Polysulfides in Lithium-Sulfur Batteries. *J. Mater. Chem. A* **2016**, 4, 12973-12980.
7. Liang, G.; Wu, J.; Qin, X.; Liu, M.; Li, Q.; He, Y. B.; Kim, J. K.; Li, B.; Kang, F., Ultrafine TiO_2 Decorated Carbon Nanofibers as Multifunctional Interlayer for High-Performance Lithium-Sulfur Battery. *ACS Appl. Mater. Inter.* **2016**, 8, 23105-23113.
8. Liu, M.; Li, Q.; Qin, X.; Liang, G.; Han, W.; Zhou, D.; He, Y. B.; Li, B.; Kang, F., Suppressing Self-Discharge and Shuttle Effect of Lithium-Sulfur Batteries with V_2O_5 -Decorated Carbon Nanofiber Interlayer. *Small* **2017**, 13, 1613-6810.
9. Xiao, Z.; Yang, Z.; Wang, L.; Nie, H.; Zhong, M.; Lai, Q.; Xu, X.; Zhang, L.; Huang, S., A Lightweight TiO_2 /Graphene Interlayer, Applied as a Highly Effective Polysulfide Absorbent for Fast, Long-Life Lithium-Sulfur Batteries. *Adv. Mater.* **2015**, 27, 2891-2898.
10. Niu, S.; Lv, W.; Zhou, G.; Shi, H.; Qin, X.; Zheng, C.; Zhou, T.; Luo, C.; Deng, Y.; Li, B.; Kang, F.; Yang, Q.-H., Electrostatic-Spraying an Ultrathin, Multifunctional and Compact Coating onto a Cathode for a Long-Life and High-Rate Lithium-Sulfur Battery. *Nano Energy* **2016**, 30, 138-145.